Coprecipitation of Carrier-free 96Tc04- with Teraphenylarsonium Perchlorate

VI. 4 Coprecipitation of Carrier-free $^{96}TcO_4^-$ with Teraphenylarsonium Perchlorate

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Pertechnetate which is an important starting compound in the syntheses of ^{99m}Tc-labeled radiopharmaceuticals can be extracted with tetraphenylarsonium chloride(TPAC) into the organic phase.[1,2] In the perchlorate solution, however, its reaction with TPAC has not yet been investigated. In the present study, coprecipitation behavior of carrier-free $^{96}TcO_4^-$ with tetraphenylarsonium perchlorate(ϕ_4AsClO_4) has been examined in detail. Furthermore, production of ^{96}Tc by the $^{93}Nb(\alpha,n)^{96}Tc$ reaction will be discussed. Tc-96($T_{1/2}$ = 4.35 d) is more effective than ^{99m}Tc as a tracer of technetium.

EXPERIMENTAL

a) Production of ^{96}Tc. Tc-96 was produced by the $^{93}Nb(\alpha,n)^{96}Tc$ reaction. The stacked foils in which Ag(105 mg/cm^2) and Nb (8.6 mg/cm^2) foil were placed between Al foils were bombarded with 35 MeV α particles (av. 5 μA) for 2 h using an AVF cyclotron at the Cyclotron and Radioisotope Center, Tohoku University. The silver foil was employed for the simultaneous production of ^{111}In by the $^{109}Ag(\alpha,2n)^{111}In$ reaction.

b) Preparation of carrier-free $^{96}TcO_4^-$. The target was allowed to stand for 2 or 3 days after bombardment in order to decay out the short-lived radioisotopes. The niobium foils were dissolved in HNO_3 and HF. On addition of 4 M NaOH the niobium was precipitated as hydroxide, while a large portion of the ^{96}Tc remains in the solution. The filtrate was then passed through a Dowex 1-X8 column (OH-form, 8ϕ x 30 mm). After ^{96}Tc other than the pertechnetate and the fluoride was eliminated by washing with 0.1 M NaOH, the carrier-free $^{96}TcO_4^-$ could be eluted from the column with 2 M $HClO_4$ solution.

c) Coprecipitation of $^{96}TcO_4^-$ with tetraphenylarsonium perchlorate. To a $^{96}TcO_4^-$ solution a TPAC solution was added. The precipitate was allowed to stand for 1 h at a constant temperature and then filtered. The radioactivity of each fraction was measured with a Ge(Li) detector connected to a TN4000 data processing system or a well-type NaI(Tl) detector connected to a single channel PHA.

RESULTS AND DISCUSSION

a) Yield of ^{96}Tc. First, the yield of ^{96}Tc was determined as a function of α particle energy. The yields of ^{96}Tc, ^{95}Tc($T_{1/2}$ = 20 h) and ^{95m}Tc($T_{1/2}$ = 61.2 d) at the end of bombardment are summarized in Table I. The ^{96}Tc-yield was evaluated after complete decay of ^{96m}Tc($T_{1/2}$ = 51.5 m) and then it was

extrapolated to the end of bombardment. No detectable amounts of radioactivities other than the Tc isotopes were found in the final $^{96}TcO_4^-$ solution.
b) Coprecipitation of carrier-free $^{96}TcO_4^-$ with tetraphenylarsonium perchlorate. The coprecipitation yield of $^{96}TcO_4^-$ is definitely affected with the concentration of TPAC, but it is independent of pH and the standing time of the precipitate.

In order to explain these features, the following equilibrium reaction may be assumed:

$$\phi_4AsClO_4 + TcO_4^- \overset{K}{\rightleftarrows} \phi_4AsTcO_4 + ClO_4^- \qquad (1)$$

Since the concentration of TPAC is in a large excess over that of TcO_4^- under the experimental conditions and TPAC precipitates quantitatively with perchlorate[3], a plot of $\log([\phi_4AsTcO_4]_p/[TcO_4^-]_s)$ against $\log([\phi_4AsClO_4]_p/[ClO_4^-]_s)$ should give a straight line of slope 1 and the equilibrium constant K can be determined from the intercept. Figure 1 shows the plot at 25°C. Thus, the equilibrium constants are found to be 2.6(10°C), 3.3(25°C) and 3.5(40°C).

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References

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Table I. YIELD AT EOB ($\mu Ci/\mu A \cdot hr/(mg/cm^2)$)

E_α (MeV)	^{96}Tc*	^{95}Tc	^{95m}Tc
19.7	.51	3.9	.019
17.7	.81	1.8	.014
15.6	.86	.13	.005

* $^{93}Nb(\alpha,n)^{96}Tc + {}^{93}Nb(\alpha,n)^{96m}Tc \xrightarrow{IT} {}^{96}Tc$

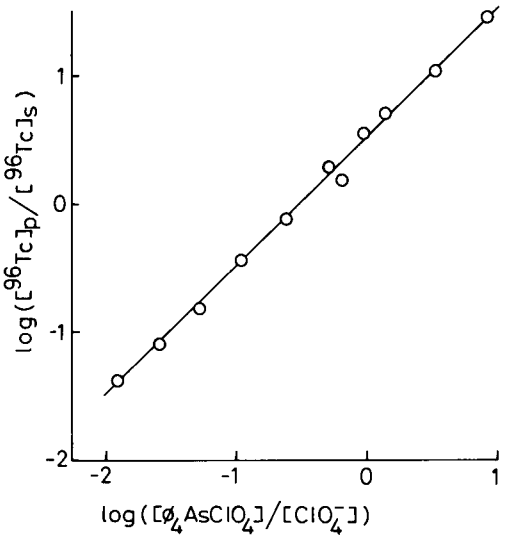

Fig. 1. $\log([\phi_4AsTcO_4]_p/[TcO_4^-]_s)$ *vs.* $\log([\phi_4AsClO_4]_p/[ClO_4^-]_s)$ (25°)